# 3 ENVIRONMENTAL RADON, THORON AND RELATED AEROSOLS

## 3.1 OVERVIEW

Alfred J. Cavallo

The 1995 EML Radon Program was active in the following areas:

***Quality Assurance Activities***

EML, with its extremely stable, NIST-traceable calibrated pulse ionization chambers and its expertise in aerosols and radon progeny measurements, has long been a reference laboratory for radon and thoron measurements. It is a regional intercomparison center for radon, thoron and progeny measurements in the International Radon Metrology Program (IRMP). As a participant in this program, it is one of three regional reference laboratories in the International Intercalibration and Intercomparison Program for Radon, Thoron and Daughter Measuring Equipment (IIIP) of the International Atomic Energy Agency (IAEA). The other two are the NRPB (National Radiation Protection Board), Chilton, UK, and the ARL (Australian Radiation Laboratory), Melbourne, Australia. It has also served as the Quality Control/Quality Assurance Center for the DOE OHER (Office of Health and Environmental Research) Radon Research Program. This work is described in Summary Nos. 3.2 - 3.6.

***Experimental Programs***

EML's experimental investigations for 1995 occurred in three project areas as follows:

(1) Exposure/Dose Assessment Studies - The risk coefficient for lung cancer due to exposure to radon progeny is based on epidemiology studies of several cohorts of uranium miners. The computed dose to these miners is based on an assumed activity weighted particle size distribution that may neglect important components. Using modern equipment, the atmosphere of a recently opened uranium mine (the Eagle Point Mine at Rabbit Lake in northern Saskatchewan, Canada) has been characterized to determine if the current assumptions on the particle size distribution are valid. A new and potentially important component of the aerosol in uranium mine atmospheres has been discovered. This project is described in Summary Nos. 3.7 and 3.8.

(2) Soil Gas Measurements - This project addressed the following issues: the importance of geologic settings on soil gas radon variations, the relative importance of variables such as permeability and soil temperatures in soil gas transport, and the investigation of the use of the ratio of thoron to radon in soil gas to distinguish between diffusion and advection transport. This investigation is described in Summary No. 3.9.

(3) Detector/New Techniques Development - Measurement technology for this program currently focuses on the development of small and portable devices for indoor radon, and thoron

concentration measurements, and on methods for measuring very low levels of radon gas. This work is described in Summary No. 3.10.

***Quality Assurance Activities***

## 3.2 NATIONAL RADON GAS INTERCOMPARISON

Isabel M. Fisenne, Andreas C. George and Pamela M. Perry

EML is the USDOE quality assurance center for radon gas and progeny measurements. A program of radon gas intercomparisons began in 1981. The Office of Energy Research, Office of Health and Environmental Research (OHER), funded major programs focused on the health effects of indoor radon. OHER recognized the need for a quality control program to establish the validity of the data obtained through its programs. Thus, OHER mandated the participation of its radon contractors in appropriate EML radon and radon progeny intercomparisons.

The 27th EML Radon Gas Intercomparison was held in April, 1995. The participating groups included three U. S. federal laboratories, one national laboratory, three state laboratories, six universities and ten commercial vendors. There were six foreign participants with three Canadian institutions and one each from Argentina, the Czech Republic and Switzerland.

For the first time, the data summary was decoded to identify the participants and to make this quality assurance information readily available to interested parties. Of the 30 participants, 3 groups were outside $\pm$10% of the EML pulse ionization chamber value of 1040 $\pm$ 20 Bq $^{222}$Rn m$^{-3}$.

An EML report documenting the results of the 1994 radon gas intercomparisons has been issued. (Fisenne, 1995).

### Reference

Fisenne, I.M., A.C. George, P.M. Perry and H.W. Keller
"The April 1994 and October 1994 Radon Intercomparisons at EML"
USDOE Report EML-568, October (1995)

## 3.3 RADON AND RADON PROGENY MEASUREMENT WORKSHOP

Andreas C. George and Keng Wu Tu

A workshop was held at EML November 15-22, 1995 to discuss, intercompare and evaluate radon and radon progeny measurements made in Japan for the assessment of the radiation exposure of the general public. There were ten participants from Japan representing the following laboratories and universities.

- Advanced Research Center for Science and Engineering
- National Institute of Radiological Sciences
- Power Reactor and Nuclear Fuel Development Corporation
- Institute of Environmental Sciences
- Japan Chemical Analysis Center
- Science University of Tokyo
- Research Reactor Institute of Kyoto University
- School of Engineering, Nagoya University
- Gifu College of Medical Technology

Passive integrating monitors consisting of activated carbon collectors, solid state electrostatic collector monitors and nuclear track monitors were exposed in the EML radon test facility under different conditions of radon concentration. Current ionization chambers were tested by three groups representing primary radon test laboratories in Japan. Scintillation cell monitors were tested and evaluated by two participants. Several continuous potential alpha energy concentration (PAEC) monitors were tested under different conditions of radon progeny, airborne particle concentrations and particle sizes. The concentrations of individual radon progeny and PAEC were measured by two participants using alpha spectrometry to count the radioactivity collected for five minutes on membrane filters. At first, both alpha spectrometric measurements deviated from the actual values due to counting efficiency differences and undetermined overlapping of $^{214}$Po into the $^{218}$Po energy region. The results of the continuous PAEC monitors indicate radon progeny losses by plateout when the particle concentration during testing was less than 5,000 particles $cm^{-3}$.

The most significant accomplishment during this intercomparison was the intercalibration of the ionization chambers currently used in Japan for their primary radon measurement methodology. The workshop was very productive in terms of assuring standardization of the instruments and methods for measuring radon and radon progeny on an international level.

## 3.4 INTERCOMPARISON OF INSTRUMENTS AND METHODS FOR MEASURING RADON AND RADON PROGENY

Andreas C. George, Keng Wu Tu, Earl O. Knutson and Isabel M. Fisenne

An intercomparison exercise for radon and radon progeny instruments and methods was held at EML from April 28 to May 10, 1995. The intercomparison was conducted in the new 30 $m^3$ radon test facility in accordance with OHER requirements and as part of the International Radon Metrology Program (IRMP). EML, as a reference laboratory for North America and Canada, conducts this intercomparison annually.

Instruments that measure radon with open face and diffusion barrier activated carbon collectors were sent by 13 participants, while 10 sent nuclear alpha track detectors, 9 with short-term and long-term electret/ionization chambers, and 13 sent active and passive commercial electronic continuous monitors. For radon progeny, there were four participants that came in person to take part in the grab sampling methodology for measuring individual radon progeny and the potential alpha energy concentration (PAEC). There were also ten participants with continuous and integrating commercial electronic instruments for PAEC. The results indicate that all the tested instruments that measure radon did well. All instruments and methods for measuring radon progeny by grab sampling techniques performed well. However, when the particle concentration is less than 5,000 $cm^{-1}$, most of the continuous commercial instruments for measuring PAEC or working level (WL) appear to underestimate the potential risk from radon progeny.

The IRMP provides technical training and advice on the use of state-of-the-art instrumentation for radon, thoron and progeny surveys and for scientific research. The IRMP is involved in the IAEA/CEC Coordinated Research Program on Radon in the Human Environment which involves more than 60 nations worldwide. The main purpose of the IRMP intercomparisons is to ensure consistency in the radon measurements on a global scale.

In the last two years, EML has been involved in preparing the main portion of a document on the design criteria and operational characteristics of radon and radon progeny calibration facilities for the IRMP. Information was collected on radon test facilities operating worldwide. The purpose of this effort was to document the characteristics of different radon test facilities so that the member nations could have easy access for calibration services and for their research needs. An inventory of more than 40 radon test facilities representing primary, secondary and field centers has been compiled and submitted to the IAEA for inclusion in the final document.

### Reference

George, A.C., E.O.Knutson, K.W. Tu and I. M. Fisenne
"Intercomparison of Active, Passive and Continuous Instruments for Radon and Radon Progeny"
USDOE Report EML-577, in press

## 3.5 COORDINATED RESEARCH AND CONSULTATIONS IN INSTRUMENT DEVELOPMENT EVALUATION

Andreas C. George

Instrument development for the assessment of the health risk from radon, thoron and their progeny continues in commercial and monitoring organizations. EML provides its test facilities for the evaluation, upgrading and refinement of instruments to insure good measurement quality. QA radon exposures were conducted with new scintillation cells for the U.S. EPA Risk Management Research Laboratory, the NY State Department of Environmental Conservation, the Pennsylvania Department of Environmental Resources and the NJ Department of Environmental Protection.

Prototype liquid scintillation type activated carbon collectors for radon were calibrated and evaluated for the Radon Testing Corporation of America and Teledyne Isotopes, Inc. Nuclear alpha track detectors were evaluated for several Japanese research laboratories. QA work was conducted to evaluate the primary methods for radon measurements used in Japan. Also, pulse ionization chambers were tested for the Oak Ridge National Laboratory (ORNL) Health Sciences Division. EML provided its radon facilities to the International Academy of Hi-Tech Services for tests on radon progeny dynamics such as attachment, surface deposition and particle size.

As a member of the DOE Radiological Control Coordinating Committee's (RCCC) Radon Subcommittee, Andreas George contributed to the final report on the "Occupational Exposure to Radon, Thoron and their Progeny." Such occupational exposure involves about 3,000 workers. The final document provides an overview of national and international guidance and regulations for occupational exposure to radon and thoron. This Subcommittee makes recommendations for the proper use of instruments and offers a comprehensive program at DOE sites in the assessment, control and recording of occupational exposure.

Andreas George also gave the "Technical Keynote Address" to the American Association of Radon Scientists and Technologists at the 1995 International Radon Symposium in Nashville, Tennessee, on the intercomparison and performance of different instruments for the measurement of radon, thoron and their progeny.

At the request of the U.S. Department of Housing and Urban Development (HUD), Andreas George reviewed the report, "Radon Testing Protocols and Measurement Devices for HUD Assisted Housing." In addition, Mr. George served as a consultant to the American Water Works Association on the impact of water-borne radon to human health during showering.

## 3.6 INTERNATIONAL INTERCOMPARISON MEASUREMENTS OF RADON PROGENY PARTICLE SIZE DISTRIBUTION

Keng-Wu Tu, Earl. O. Knutson and Alfred J. Cavallo

EML hosted an international intercomparison of measurements of radon progeny particle size distribution during the week of June 12-15, 1995. It is important that progeny particle size distribution be accurately measured because the dose to various portions of the lung is dependent on the size distribution of the particle inhaled. The purpose was to reaffirm quality control procedures and to improve communications among the different groups making the measurements. Participants included three foreign and three domestic laboratories: AEA Technology, the United Kingdom; Isotope Laboratory (IL), University of Gottingen, Germany; Australian Radiation Laboratory, Australia; U.S. Bureau of Mines, Denver Research Center; Clarkson University, Potsdam, NY; and EML, the host laboratory. The participants from AEA Technology, the Australian Radiation Laboratory, and the U.S. Bureau of Mines used the screen diffusion battery technique for this exercise, while the others chose the low pressure impaction methods for this test.

Twenty measurements were made at seven controlled particle size distributions at given radon concentrations. All the participants took samples nearly simultaneously for 5 to 15 minutes. All the samples were alpha counted except Isotope Laboratory's which were determined with a NaI detector for the gamma radioactivity from $^{214}$Pb.

At present, data has been received from three groups: AEA Technology used six screen diffusion batteries in parallel and sampled at 6.5 lpm; Isotope Laboratory used a low-pressure cascade impactor (Berner, Model LP130), which consists of eight stages and a backup filter, sampling at 30 lpm; and EML used a micro-orifice uniform deposit impactor (MOUDI, MSP Corporation, MN) which also consists of eight stages and a backup filter, sampling at 30 lpm. A graded screen array (GSA) was also used by EML to monitor the ultra-fine particles in the aerosol.

Because the Isotope Laboratory group determined activity size distributions based on gamma radioactivity of $^{214}$Pb only, we chose this progeny size for the comparison. Table 3.1 lists typical results. Most of the data obtained from the two impactors agreed quite well. The diffusion battery system provided broader size spectra for the particles with diameters greater than 100 nm. For the bimodal size distribution particles, only the MOUDI observed minor modes with diameters less than 150 nm.

## TABLE 3.1

## Comparison of $^{214}Pb$ Particle Size Distributions from the EML MOUDI, IL BERNER Imactor and AEA Diffusion Battery System.

| | | | | EML | | Isotope Lab | | AEA | |
|---|---|---|---|---|---|---|---|---|---|
| Run # | Rn Conc Bq $m^{-3}$ | CNC++ 1000 $cm^{-3}$ | Dc+ nm | AGMD* nm | GSD** | AGMD* nm | GSD** | AGMD* nm | GSD** |
| 121447 | 2230 | 7 | 80 | 56 | 1.58 | 93 | 2.86 | 69 | 1.98 |
| 121600 | 1991 | 7 | | 67 | 2.67 | 163 | 2.62 | -- | -- |
| 121700 | 1784 | 8 | | 79 | 1.7 | 85 | 3.13 | -- | -- |
| 130900 | 1894 | 8 | 90 | -- | -- | 86 | 1.68 | 281 | 3.7 |
| 131000 | 1868 | 14 | | 84 | 1.61 | 89 | 1.76 | 119 | 2.67 |
| 131110 | 1600 | 16 | | 84 | 1.6 | 92 | 1.64 | 635 | 7.58 |
| 131405 | 2156 | 17 | 165 | 174 | 1.37 | 172 | 1.54 | 446 | 3.43 |
| 131505 | 1631 | 18 | | 158 | 1.56 | 172 | 1.54 | 485 | 1.94 |
| 131610 | 1233 | 20 | | 167 | 1.6 | 177 | 1.5 | 504 | 1.46 |
| 140930 | 870 | 6.7 | 395 | 374 | 1.31 | 356 | 1.89 | 504 | 1.27 |
| 141040 | 939 | 7.7 | | 375 | 1.3 | 380 | 1.63 | 504 | 1.27 |
| 141200 | 776 | 8.4 | | 393 | 1.34 | 395 | 1.90 | 494 | 1.59 |
| 141400 | 896 | 16 | 160 & 365 Bimodal | 103<br>375 | 1.33<br>1.32 | --<br>291 | --<br>2.17 | --<br>480 | --<br>1.87 |
| 141520 | 871 | 14 | 160 & 365 Bimodal | 104<br>374 | 1.36<br>1.34 | --<br>344 | --<br>1.63 | --<br>451 | --<br>2.23 |
| 141620 | 902 | 15 | 160 & 365 Bimodal | 83<br>374 | 1.78<br>1.35 | --<br>289 | --<br>1.88 | --<br>497 | --<br>1.51 |
| 150900 | 2009 | 10 | 70 & 400 Bimodal | 95<br>377 | 1.38<br>1.32 | --<br>323 | --<br>1.81 | --<br>440 | --<br>2.81 |
| 151000 | 1694 | 10 | 70 & 400 Bimodal | 110<br>386 | 1.33<br>1.37 | --<br>421 | --<br>1.85 | --<br>502 | --<br>1.45 |
| 151110 | 1290 | 10 | 70 & 400 Bimodal | 100<br>375 | 1.34<br>1.32 | --<br>383 | --<br>1.74 | --<br>505 | --<br>1.38 |
| 151330 | 1876 | 11 | 1200 | 1277 | 1.32 | 1327 | 1.18 | -- | -- |
| 151420 | 2050 | 10 | | 1187 | 1.41 | 1373 | 1.18 | -- | -- |

* AGMD: Activity-weighted geometric mean diameter
** GSD: Geometric standard deviation
\+ Dc: Controlled particle modal diameter
++ CNC: Particle number concentration with TSI 3025 condensation nuclei counter

*Experimental Programs*

## 3.7 EXPOSURE/DOSE ASSESSMENT

Alfred J. Cavallo, Adam R. Hutter and Keng-Wu Tu

It is critical that the calculation of the human risk coefficient for radon exposure be based on as sound a scientific basis as possible, given the limitations that are inherent in its derivation. This risk coefficient is used to set limits both on occupational and domestic radon levels. Since the uranium mining and processing industry is now quite small, the impact that domestic exposure limits has is actually far more important. Domestic radon exposure is caused by radon contaminated soil gas that is drawn into homes due to normal indoor-outdoor pressure differentials. It was recognized only in the early 1980s that this exposure accounts for more than half of the average estimated dose equivalent in the U.S. population (NCRP, 1987). Thus, the estimate of the radon risk coefficient has been examined carefully by the Committee on the Biological Effects of Ionizing Radiation (BEIR IV, 1988). The current guideline of 4 pCi $L^{-1}$ for radon exposure promulgated by the U.S. Environmental Protection Agency is based in large part on the radon risk coefficient given this group.

This risk coefficient is determined from long-term epidemiological studies of several different groups of miners. The dose received by these miners depends not only on the magnitude of their exposure to radon progeny, but also on the size of the particles to which the radon progeny were attached, the so-called activity weighted particle distribution, as well as on breathing patterns; these factors govern the deposition of particles in the tracheobronchial region of the lung (believed to be the portion of the lung most at risk). Figure 3.1 (ICRP, 1994).shows deposition for a breathing rate corresponding to light exercise, the most important miner breathing pattern. Note that the deposition as a function of particle size is virtually flat from about 150 nm to above 20,000 nm; as long as the inhaled particles were in this range, the details of their distribution have no impact on total dose. Figure 3.2 (ICRP, 1994) illustrates deposition as a function of particle size for a breathing rate corresponding to heavy exercise. In this case, both the magnitude and the shape of the deposition changes compared to the lower breathing rate; not only is the deposition higher overall, but also large particles (those with diameters greater than 1000 nm) have a much higher deposition rate.

Current risk coefficient estimates are based on an assumed particle size distribution in the range of 150 to 250 nm (NRC, 1991). However, based on previous work in an experimental uranium mine (Knutson and Tu, 1995), it was felt that this may neglect a significant fraction of the activity attached to particles with a diameter greater than 1000 nm. To resolve this question, a field trip to an active uranium mine was organized. The aim of this trip was to characterize the mine atmosphere as completely as possible, with particular attention to those locations that might duplicate conditions in old mines. The uranium mine selected was the Eagle Point Mine at Rabbit Lake in northern Saskatchewan, Canada, owned and operated by the Cameco Corporation. (We are especially grateful for the complete cooperation and help provided by Cameco employees.)
Equipment used included:

- MOUDI (Micro Orifice Uniform Deposit Impactor)
- Graded Screen Array
- Associated counting equipment (photomultiplier tubes, computers)
- Alphaguard (radon measurements)
- B&K Multigas Spectrometer ($CO_2$, CO, $H_2O$)
- Met 1 (Particle size distributions, particle diameter >500 nm)
- Electrical Aerosol Analyzer (particle size distributions)
- CNC (condensation nuclei counter, particle concentration)

Conditions in the mine were particularly challenging, especially for equipment not specifically designed for field use. Temperature in the mine was about 5° C (outside temperatures were -15° C) with a relative humidity of 80 - 100 %. Water dripped from the mine roof and accumulated on the floor, causing electrical short circuits in some of the equipment. The high ventilation rate needed to keep radon levels acceptably low combined with the high humidity and low temperatures to make the working environment physically stressful in spite of many layers of clothing. Some rough idea of the conditions experienced in this mine can be obtained by examining Figure 3.3, which shows an activity-weighted particle size distribution sample being taken using the MOUDI impactor at a site of active drilling.

Radon levels in the mine ranged from 15,000 Bq $m^{-3}$ (400 pCi $L^{-1}$, normally off limits for miners) at the top of a working stope (a position at which the ore is being extracted) to 567 Bq $m^{-3}$ (15 pCi $L^{-1}$) at a drilling site. A condensation nuclear counter indicated that particle number concentration was always greater than 70,000 $cm^{-3}$ everywhere in the mine, even in areas with no mining or other activity. Carbon dioxide and carbon monoxide levels were about four times the levels normally found in indoor air; filters in the overnight samplers were black with what appeared to be soot.

A typical activity weighted particle size distribution for the PAEC, shown in Figure 3.4, was centered over the range of 50-100 nm, with virtually no activity outside this range. Virtually all of the activity is due to the short-lived radon progeny $^{218}Po$, which is consistent with the high ventilation rate.

At first, we attributed the aerosol characteristics of the mine atmosphere to the underground use of diesel powered ore carriers and jeeps. However, given the high ventilation rates for the mine (1.3 million cubic feet per minute), it seemed completely unreasonable that a small number of diesel engines could possibly create the high particle concentrations and the high CO and $CO_2$ levels. The particle size distribution, moreover, was not at all typical of what is observed with diesel engines, which is usually centered in the range of 150 to 250 nm (NRC, 1991).

We then realized that the fresh air pumped underground was heated by burning propane in the airstream at a rate of about 25 lpm with the combustion products being carried into the mine with the fresh air. (Outside temperature, as noted above, was significantly colder than in the mine.) Although propane is a clean-burning fuel, a minute amount of fine carbon aerosol is formed during combustion; formation of a fine aerosol during atmospheric burning of propane or natural gas is a well known phenomenon. This also accounts for the higher CO and $CO_2$ levels observed in the mine

atmosphere, as well as the blackening of our filters.

Discussions with a mining engineer at the Eagle Point Mine revealed that in his experience this heating technique has, for the last 15 years, been a common practice in Canadian mines; there is no other practical way to heat the large volumes of air needed for mine ventilation.

The impact these findings will have on estimates of the risk coefficient is difficult to quantify since the tracheobronchial deposition changes rapidly in this size range, and assumptions on particle growth must be examined carefully. However, during the heating season, an increase in dose per unit of exposure of a factor of 1.5 to 2 seems possible. This finding may have important implications for previous epidemiology studies of Canadian miners. In addition, it is clear that instruments that monitor radon progeny must be calibrated over a very wide range of particle diameters if they are to measure airborne radioactivity accurately. Additional mine studies are planned to advance our knowledge of the aerosol characteristics in contemporary and earlier uranium mine atmosphere and its impact on the estimate of the human risk factor to radon exposure.

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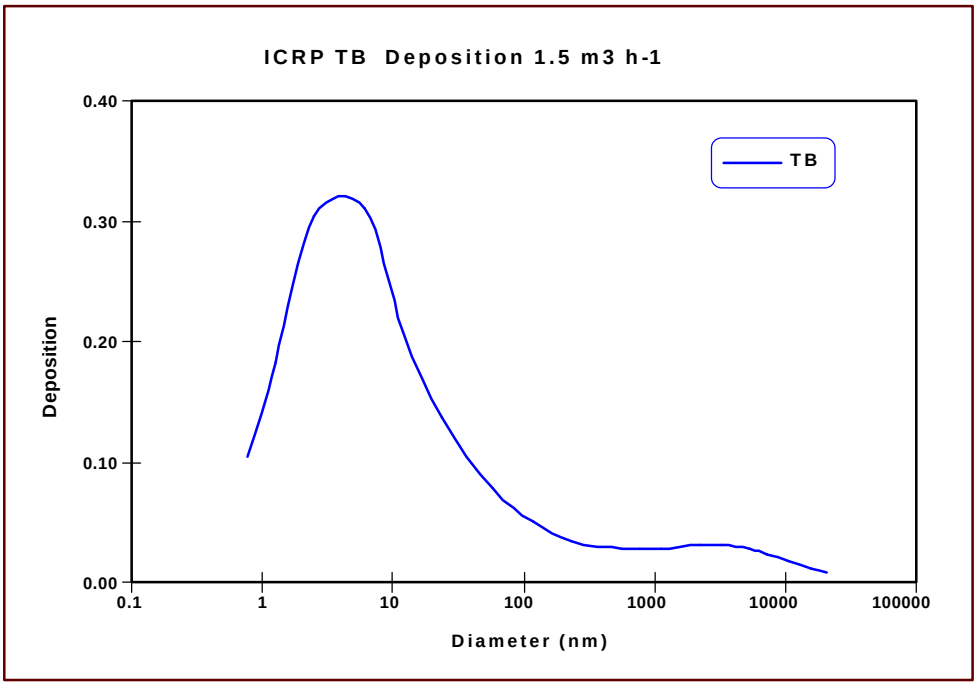


**Figure 3.1** Tracheobronchial deposition as a function of particle diameter for light exercise (adult male, breathing rate, 1.5 $m^3$ $h^{-1}$, normal nose breathing). It is assumed that miners breathed at this rate 75 percent of the time that they spent at the ore face, and all of the time that they spent working in other areas of the mine (ICRP 1994; NRC, 1991).

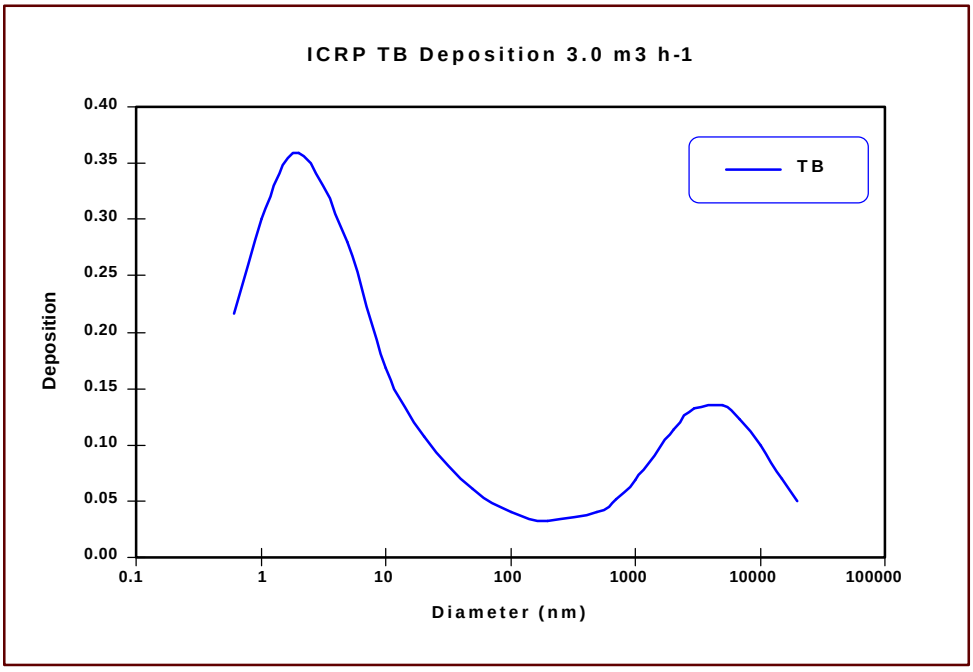


**Figure 3.2** Tracheobronchial deposition as a function of particle diameter for heavy exercise (adult male, 3 $m^3 h^{-1}$, normal nose augmented breathing). It is assumed that miners breathed at this rate 25 percent of the time that they spent at the ore face (ICRP 1994; NRC, 1991).

**Figure 3.3** Taking a sample with the MOUDI to measure the activity weighted particle size distribution at an active drilling site in the Eagle Point mine 260 meters below the surface.

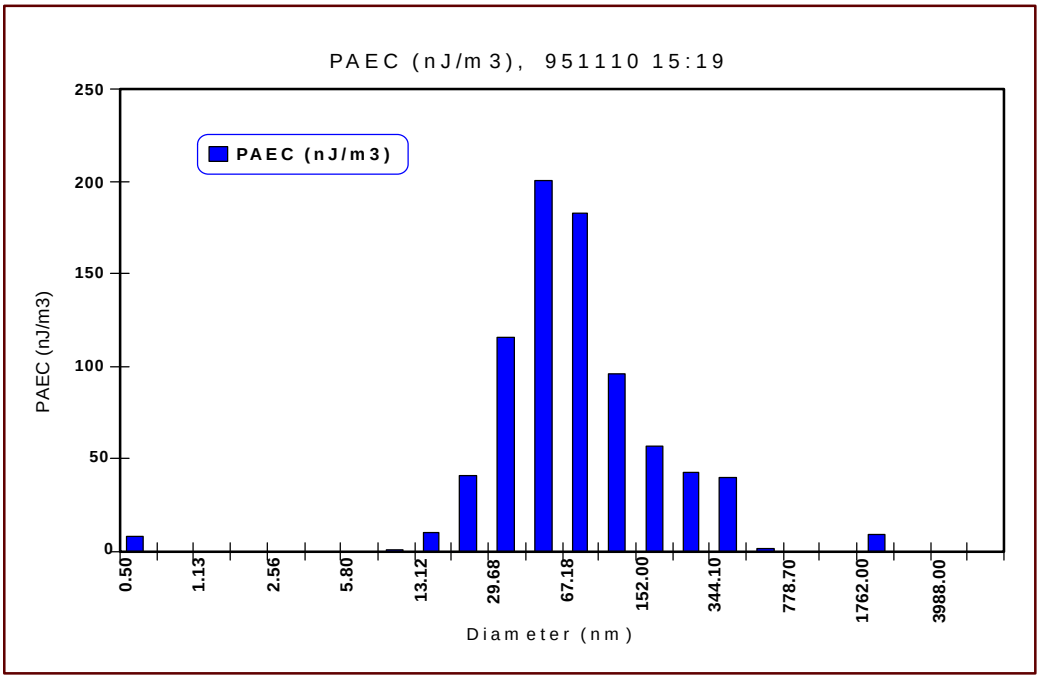


**Figure 3.4** PAEC (potential alpha energy concentration, nJ/m-3) as a function of particle diameter(nm). This data was taken on November 10, 1995, at 15:19 in the bolt storage area (located in an alcove off the main decline) about 90 meters below the surface.

## 3.8 ELECTRON MICROSCOPIC CHARACTERIZATION OF AEROSOLS IN THE RABBIT LAKE URANIUM MINE

Lambros Kromidas, Adam R. Hutter, Alfred J. Cavallo, Keng-Wu Tu, and Peter Roiz

Three aerosol samples analyzed with the electron microscope were collected about twelve inches above the floor of a uranium mine (see Summary No. 3.7) using a radon progeny monitor Pylon AB-5 with an AEP-47 Alpha Detection Assembly. The samples were collected on 10.5 $cm^2$ polycarbonate Nuclepore filters (0.8 $\mu$m pore size). The first sample was retrieved on November 8, 1995 in an isolated location (120 m level) in the mine where very little vehicular activity was taking place. This specimen was accumulated over a period of 24 hours and sampled 1,440 L of air. The second (11/11/95) and third (11/12/95) specimens were collected at the 90 m level, referred to as the bolt storage area. This area was located on the main decline and was characterized by high vehicular activity. The 11/11/95 sample filtered 2,820 L of air over an interval of 47 hours. The 11/12/95 collection sampled 1,740 L of air over an interval of 29 hours.

The filters were sectioned, and a representative portion of each was coated with 10 nm of platinum by a Denton Vacuum Evaporator, DV-502A (Denton Vacuum Inc., Cherry Hill, NJ). Backscatter microscopy of the aerosols was performed by a Robinson Detector (ETPSEMRA, Sydney, Australia) and energy dispersive X-ray microanalysis was performed by the Princeton Gamma-Tech Energy Dispersive Spectrometer, Model 4200, PGT, Princeton, NJ with a two position, light element, lithium drifted silicon x-ray detector.

Results show that the filters contain two major types of particulate aerosols, namely soot and soil. All three samples contained an abundance of soot particles ranging in size from the submicron range to large concatenated clusters many microns in size. Due to their abundance and clustering, obtaining concentration density and size distribution was not possible. X-ray microanalysis of the soot did not reveal any detectable contaminants.

The 11/8/95 sample indicated the concentration of soil particles to be 3,806 particles $L^{-1}$ of air. The average longest dimension of these particles was 1.2 $\mu$m. The concentration of soil particles in the 11/12/95 sample was estimated to be 2,870 particles $L^{-1}$ of air. Their average longest dimension was 1.5 $\mu$m. These measurements agree with those from the MET-1 particle counter, which indicated average particle concentration of less than 5000 particles per liter for particles with a diameter greater than 0.5 $\mu$m everywhere in the mine.

## 3.9 SOIL GAS $^{220}$Rn AND $^{222}$Rn

Adam R. Hutter

The soil gas Rn project culminated in 1995 with a publication in Health Physics (Hutter, 1995), a paper accepted for publication in Environment International (Hutter, in press), presentations at the Sixth International Symposium on the Natural Radiation Environment (Montreal, Canada, June 1995) and the International Radon Symposium (Nashville, Tennessee, September 27-29, 1995) and organizing and hosting the Sixth International Radon Metrology Program Intercomparison Test and Workshop at EML, June 12-15, 1995. The project addressed issues concerning the importance of geologic settings on soil gas Rn variations; the relative importance of variables affecting soil gas transport; the determination of whether diffusion is the dominant mechanism of soil gas transport, or if other mechanisms may cause unexpected variations in $^{222}$Rn concentrations; the investigation of long-term trends and the associated controlling parameters of variations in soil gas $^{220}$Rn; and the investigation of the use of $^{220}$Rn/$^{222}$Rn ratio trends to distinguish between steady-state diffusion-dominant migration of soil gas versus advection-dominant.

The project entailed field sampling at three New Jersey sites over a three-year period and resulted in over 1200 soil gas $^{222}$Rn measurements, 900 $^{220}$Rn soil gas measurements, and 900 soil permeability measurements. The major findings of the project include:

(1) A one-time sample for the determination of soil gas $^{222}$Rn and $^{220}$Rn concentrations may be accurate only to an order of magnitude due to seasonal effects and the disruption of the soil structure in taking the sample.

(2) Geologic setting strongly influences soil gas Rn dynamics. At a coastal plain site, neither temporal nor spatial variations were observed. At a site near the Reading Prong, strong seasonal and spatial variations in soil gas $^{222}$Rn were observed, the timing and magnitude of which defy the widely accepted, yet frequently challenged, conventions concerning soil gas transport.

(3) Diffusive transport cannot explain the timing and magnitude of soil gas $^{222}$Rn variations, suggesting that different transport modes may dominate at different times during the year, i.e., diffusion and advection.

(4) The project resulted in the first published long-term trends of soil gas $^{220}$Rn. The highest soil gas $^{220}$Rn concentrations occur during mid-summer, coinciding with predictions from diffusion-only models. These variations are largely controlled by temperature, both outdoor and soil, with up to 96% of the variability due to temperature alone at some depths (see Figure 3.5).

(5) Permeability measurements, thought to be a major indicator of parameters controlling soil gas Rn variations, show no correlation with $^{222}$Rn or $^{220}$Rn at any of the sites. This perhaps indicates that soil permeability, as measured, does not accurately reflect parameters thought to control soil gas $^{220}$Rn and $^{222}$Rn, i.e., soil moisture and porosity.

These results give credence to using radon as a tracer as a means of understanding soil-gas transport in the vadose zone, often important in planning and monitoring remedial actions at many DOE sites.

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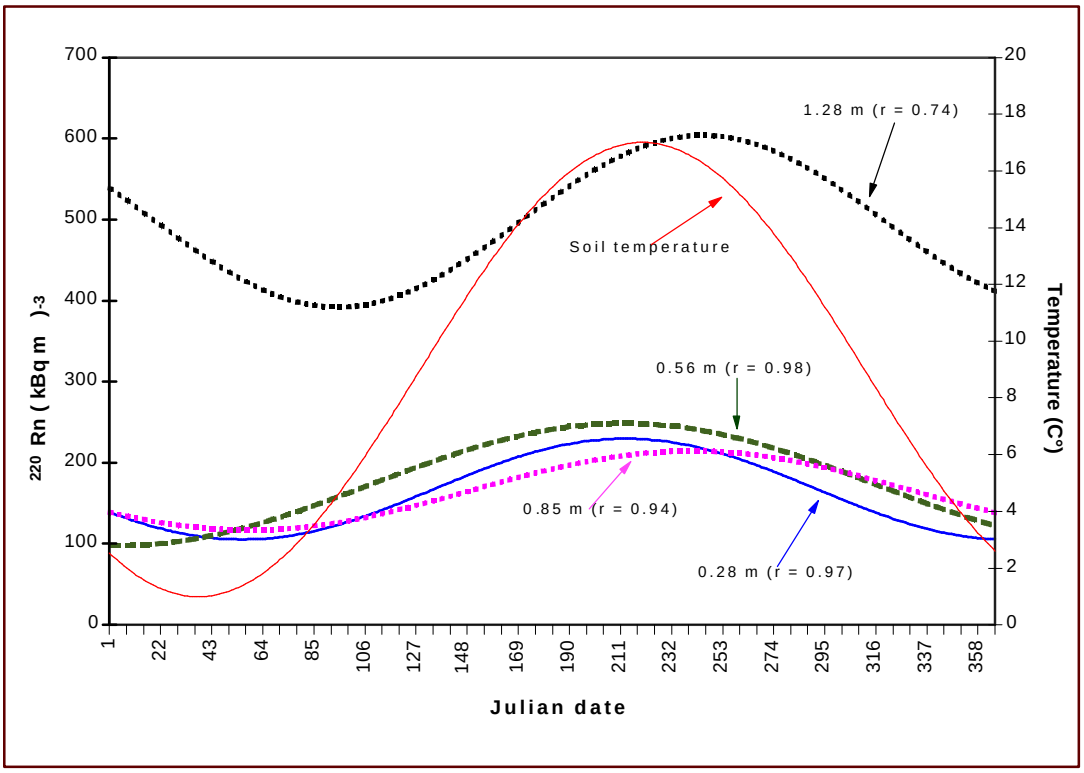


**Figure 3.5** Results of soil gas $^{220}$Rn profile from three years of data from Chester, NJ site. Soil gas $^{220}$Rn and soil temperature lines are data fit following Fourier analyses. Correlation coefficients in legend indicate the r-value between specific soil gas $^{220}$Rn depth and the modeled soil temperature.

# 3.10 SOLID PHASE SCINTILLATION COUNTING OF $^{222}$Rn USING SCINTILLATING PLASTIC BEADS

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Two commercially available plastic beaded scintillation materials are being investigated as alternatives to using 10-15 mL of toluene-based liquid scintillation cocktails for measuring $^{222}$Rn by Liquid Scintillation (LS) counting (see Summary No. 6.8). It is well known that by employing LS counting and using low background 20 mL plastic vials, an order of magnitude increase in counting sensitivity can be achieved as compared to gamma counting. For every atom of $^{222}$Rn present, three $\alpha$'s and 2 hard $\beta$'s are emitted at radioactive equilibrium providing a maximum LS count rate of 11 cpm pCi$^{-1}$ of $^{222}$Rn (assuming 100% counting efficiency).

Alpha-scintillating plastics, a polyvinyltoluene (PVT) polymer* and polyvinylbenzene (PVB) polymer+, behave similarly to liquid scintillates, producing detectable photons with a wavelength of 400-500 nm due to the interactions of $\alpha$, $\beta$ and/or $\gamma$ emitting progeny, Pb, Bi and Po. These translucent beaded scintillation materials of 100-500 $\mu$m diameter, when mixed into small amounts of activated charcoal (or other $^{222}$Rn adsorbents), should produce equivalent photon yields compared to a toluene-based liquid scintillant.

Gram amounts of Carboxen, a hydrophobic Beaded Carbon Molecular Sieve (BCMS) material<, with a $^{222}$Rn adsorptive capacity that is 2-3 times greater than any activated charcoal, were aliquoted into 20 mL low background plastic scintillation vials and exposed in the EML Radon Chamber for 48 hours at 70% humidity at a radon concentration of 51 pCi L$^{-1}$. Following exposure, 1 gram of either the PVB or PVT polymer beads was mixed into 1 gram of the BCMS material and counted in a LS analyzer. The results indicate that the PVT material yielded a counting efficiency (CE) of 3 cpm g$^{-1}$ per pCi L$^{-1}$ of $^{222}$Rn which was a factor of two greater than the observed CE using the PVB material. The CE obtained after the addition of PVT was itself a factor of 3 greater than the CE obtained when the BCMS/$^{222}$ Rn adsorbent was counted <u>without</u> the scintillating polymer beads. The exposed BCMS material by itself yielded a detectable count rate (37 cpm $\pm$ 8%) above background (16 cpm) due to Cerenkov radiation which is produced by $\beta$- particle interactions with both the BCMS material and the plastic counting vials. Both polymer beads, by themselves, did not adsorb any $^{222}$Rn, as determined from the measured count rates. When 10 mL of Perma-Fluor-V$^{++}$ (90% toluene, 10% methanol) scintillation cocktail was added to each vial containing the BCMS/$^{222}$Rn sorbent, the average CE was 15 $\pm$ 1 cpm g$^{-1}$ per pCi L$^{-1}$ of $^{222}$Rn.

Preliminary results indicate that one gram of beaded PVT polymer, when added to 1 gm of a BCMS sorbent containing adsorbed $^{222}$Rn, produces photon signals that can be measured by any LS spectrometer with about one fifth the counting efficiency of a toluene-based liquid scintillant. This reduction is most probably due to photon and alpha attenuation by the BCMS material, which is not translucent. For typical indoor radon concentrations, the combined collection/counting efficiency of 3 cpm g$^{-1}$ per pCi L$^{-1}$ provides activities well above the LLd $_{(1\ hr)}$ of 0.11 pCi (0.24 dpm) for the Packard Counter. The PVT/BCMS mix can be reused by heating at 70° C to desorb both adsorbed $^{222}$Rn and water vapor.

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< Supelco Inc., Supelco Park, Bellefonte, PA 16823
[++] Packard Instrument Co., Downers Grove, IL

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